Measurements and analysis of the upper critical field H_{c2} of underdoped and overdoped $La_{2-x}Sr_xCuO_4$ compounds

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The upper critical field H_{c2} is one of the many non conventional properties of high-T_c cuprates. It is possible that the $H_{c2}(T)$ anomalies are due to the presence of inhomogeneities in the local charge carrier density ρ of the CuO_2 planes. In order to study this point, we have prepared good quality samples of polycrystalline $La_{2-x}Sr_xCuO_4$ using the wet-chemical method, which has been demonstrated to produce samples with a good cation distribution. In particular, we have studied the temperature dependence of the upper critical field, $H_{c2}(T)$, through magnetization measurements on two samples with opposite average carrier concentration ($\rho_m = x$) and nearly equal critical temperatures, namely, $\rho_m = 0.08$ (underdoped) and $\rho_m = 0.25$ (overdoped). The results close to T_c do not follow the usual Ginzburg-Landau theory and are interpreted by a theory which takes into account the influence of the inhomogeneities.

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I. INTRODUCTION

High critical temperature superconductors (HTSC) display many non-conventional properties which remain to be explained by a concise physical picture[1, 2, 3]. This state of affairs might be due to the fact that, differently from low temperature superconductors, these materials have a large degree of intrinsic inhomogeneities. Although its origin is unknown, there are many evidences from different experiments that they do not have a homogeneous doping level[4, 5]. For instance, recent Scanning Tunneling Microscopy (STM)[6, 7, 8, 9] measurements have detected strong variations in the density of states as the tip travels over a clean and sharp surface. Also, neutron diffraction data have revealed a complex structure of the charge distribution that has become known as "stripe structure"[10]. Nuclear Quadruple Resonance (NQR)[11] and Angle Resolve Photo-Emission (ARPES)[12] have also detected inhomogeneities in the local environment and in the charge distribution.

Based on these experimental evidences we argue that this behavior and the inhomogeneities are possibly due to a phase separation transition (PST) connected with the anomalies seen at the upper pseudogap temperature. Such PST would bring the system to a disordered charge distribution state, with the formation of islands or regions of distinct doping levels[13]. Since any PST depends on the chemical mobility, this approach may shed some light on the reason why some compounds appear to be more homogeneous or, at least, do not display any gross inhomogeneity[14, 15], although the phase diagrams of cuprates seem to be universal. Therefore, we think it is possible to formulate a unified theory for the HTSC despite of their different degrees of disorder.

In this paper we explore this possibility by showing that the anomalies of the upper critical field $H_{c2}(T)$ as function of the temperature T are in agreement with charge carriers inhomogeneities in two samples with completely different average doping levels: one underdoped and other in the far overdoped region of the superconducting phase diagram. In order to achieve this goal this work is threefold: i) we prepared samples of

$La_{2-x}Sr_xCuO_4$ (LSCO), ii) we made several sets of $H_{c2}(T)$ measurements and iii) perform a theoretical interpretation of the data. The non conventional features of $H_{c2}(T)$, like a positive curvature and absence of saturation at low temperatures, are well known from many previous experiments[16, 17, 18]. We concentrat in two samples in the underdoped and overdoped doping levels, and measured $H_{c2}(T)$ to detect qualitative doping dependent behavior. We interpreted the results through a theory that takes into account the different contribution from stripes of different local charge density. These calculations are based on the Cahn-Hilliard (CH) theory of phase separation[19, 20] for compounds with $\rho_m \leq 0.20$[2, 13] and on a Gaussian charge fluctuation around the average for $\rho_m \geq 0.20$. In both cases the resulting inhomogeneous systems are studied with a Bogoliubov-deGennes approach to a disordered superconductor [13]. Indeed there are several different approaches to deal with the inhomogeneities in HTSC, like the method of Ghosal at al[21] of a local disorder in the chemical potential, Nunner et al[22] that deals with out of plane chemical disorder, and Cabo et al[23] that introduced an in plane Gaussian disorder around de average doping, to mention just a few of what can be found in the literature. So far, all these approaches succeeded in explain some HTSC features, what shows that the inhomogeneities are important, but only new and refined experiments will be able to determine the correct way to deal with them.

II. SAMPLE PREPARATION

Several polycrystalline samples belonging to the $La_{2-x}Sr_xCuO_4$ system were prepared by the wet-chemical method according to reference[24, 25]. Pure (99,99%) oxide and carbonate compounds, namely La_2O_3, CuO, and $SrCO_3$ were dried at 150^0C and weighted with adequate stoichiometrical proportions. The powders were dissolved into 50 ml of ultra pure acetic acid (CH_3COOH) and the final solution was dried and after heated at 900^0C during 24 hours in flowing oxygen. After that the powders were quenched at room temperature, then the mixtures were reground and pressed into pellets. Finally the pre-sintered samples were sintered at 1050^0C during 50 hours.

Fig.(1) shows the x-rays diffraction performed in both samples prepared according the wet-chemical method. The x-ray diffraction (XRD) patterns were obtained in a (XPert PRO PANalytical) powder diffractometer using CuK_α radiation ($\lambda = 1.5418\mathring{A}$). Data were collected by step-scanning mode ($20^0 \leq 2\theta \leq 90^0$) and 2 s counting time in each step at room temperature. Orthorhombic (Bmab) and tetragonal (F4mmm) structure space groups were assumed in the Rietveld analysis for the 0.08 and 0.25 strontium percent samples respectively.

Once we have characterized the sample, we have performed magnetic measurements by a SQUID magnetome-
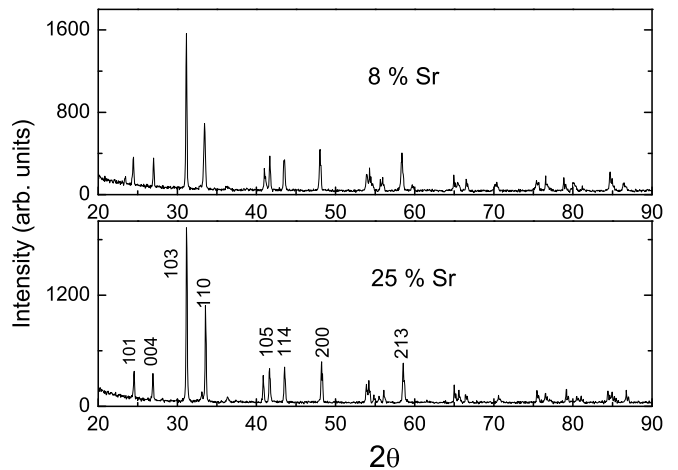

FIG. 1: X-rays diffraction spectra at room temperature for both samples used in the experiment. The principal peaks were identified according to Rietveld analysis.

ter, as described in the next section.
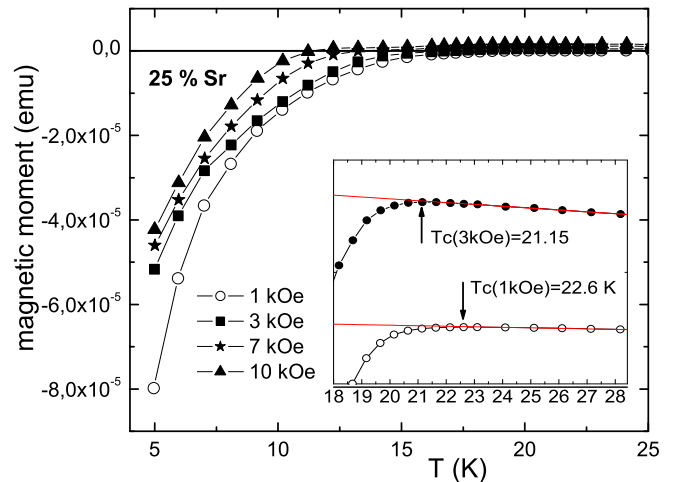
III. EXPERIMENTAL PROCEDURE

FIG. 2: (Color on line) The zero-field cooling magnetic moment at some applied magnetic fields as a function of temperature for the sample with 25% of strontium. The insert shows how the values of T_c were determined for two values of the applied field.

Magnetization curves as a function of temperature

were obtained with a SQUID magnetometer in a conventional DC mode. The measurements were performed in zero field-cooling conditions with moderate applied fields ranging between zero and two Tesla. Fig.(2) contains sets of M(T) curves for the sample $La_{1.75}Sr_{0.25}CuO_4$ (25 % of strontium) is presented. In the inset we show details of how $T_c(H)$ were obtained for two selected fields, namely 3kG and 1 kG. By taking the measured values of $T_c(H)$, we obtain the plot shown in Fig.(3), from which we can extrapolate the value of $T_c(H_{c2} = 0)$. For the present case, we obtain $T_c(H_{c2} = 0) = 24.1K$ and for the under-doped sample, we get $T_c(H_{c2} = 0) = 31.1K$. A similar set of curves was obtained for the 8% strontium sample. In this manner we got the value of $T_c(H_{c2} = 0)$ for both samples.
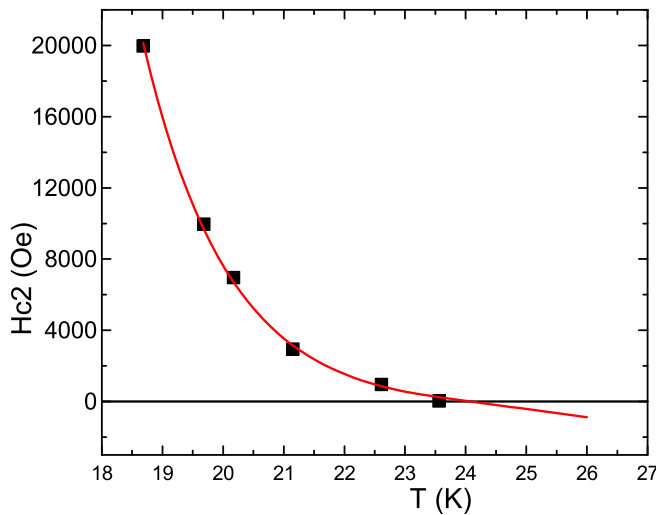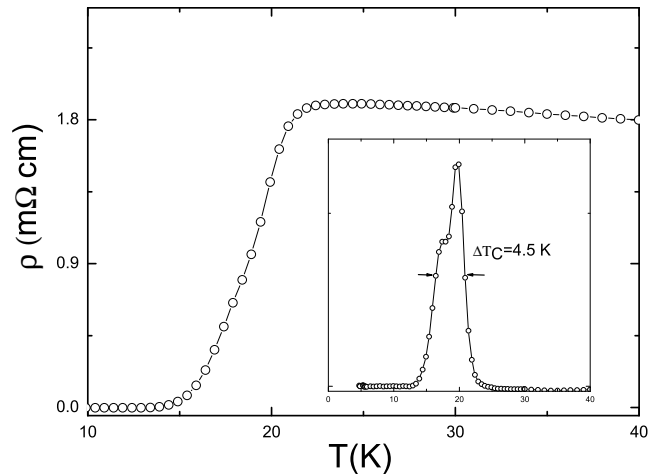

FIG. 4: Resistivity as function of temperature. The insert shows the first derivative of the data and the maximum is taken as the superconductivity transition temperature for this sample at $T_c(25\%Sr) = 19.9$K

FIG. 3: (Color on line) Values of $T_c(H)$ calculated as shown in inset of Fig.(2) (for $La_{1.75}Sr_{0.25}CuO_4$). The value of $T_c(H_{c2} = 0)$ is taken where the curve extrapolates to zero. In this case we get $T_c(H_{c2} = 0) = 24.1K$, value that will be used further on (Fig.(7)).

On the other hand, the critical temperatures $T_c(H = 0)$ for both samples were determined from many sets of resistivity data, by taking the maximum of the first derivative of the resistivity vs temperature curves, namely $T_c(8\%Sr) = 22.8$K and $T_c(25\%Sr) = 19.9$K. From these data, the widths of the superconducting transitions were estimated at half-maximum of the first derivative, as displayed in Fig.(4). For the sample with 25% of strontium the ΔT_c was about 4.5 K while in the 0.08% sample the transition was wider pointing out to the presence of strong inhomogeneities in the sample. The presence of these disorder will be important in the discussions presented in the next sections.

IV.   PST RESULTS AND DISCUSSION

As discussed in the introduction, there are many experimental evidences showing that some cuprates are highly inhomogeneous in their charge distribution while others are not, but all of them display the same phase diagram. To deal with this non trivial problem, we have introduced the idea of a PST that, depending on the mobility of the ions, can generates various degrees of decomposition. This phase segregation process can form patterns on the sample, as the stripes[10] or patchwork[6], generating islands with different values of the charge density, or can merely form small fluctuations around an average doping level. Applying a Bogoliubov-deGennes (BdG) theory to these systems we were able to calculate the local superconducting pairing amplitude at a given site "i" in a cluster "l". Thus, in our calculations, a given sample with average or mean charge density ρ_m may be composed of local regions with local densities $\rho(l)$. Here we show results on a 14×14 matrix, that is, 14 clusters on a stripe form (each stripe has 14 sites) $(l = 1 to 14)$ and 196 sites (i runs from 1 to 196).

In general, regions with larger charge densities or doping levels usually may become superconducting, and those with very low doping level are insulators and never become superconducting. This anomalous behavior is detected by the superconducting pairing amplitude $\Delta(l, T)$ which, as the temperature is decreased, starts at $T_c(l)$, increases and saturates at low temperatures. This allows us to define a *local superconducting temperature* $T_c(l)$.

Here we exhibit simulations on a square mesh 14×14 that display stripe inhomogeneities similar to the experimental results[10], but derived from a CH phase separa-
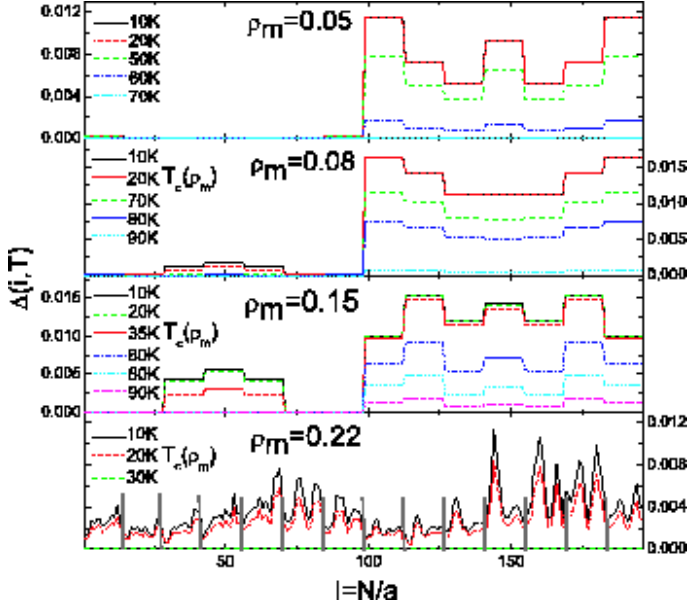
FIG. 5: (Color on line) Temperature evolution of the local pairing amplitude $\Delta(i,T)$ at each stripe on a square of 14×14, that is, with 196 sites "i", for different samples. Because of the inhomogeneities, the sites in the left have $\rho(i) \approx 0$ and the ones in the right $\rho(i) \approx 2\rho_m$. As the systems are cooled down, more regions become superconducting and they percolate at $T_c(\rho_m)$. These percolation threshold temperatures for each compound are indicated in their respective panel. The panel with $\rho = 0.22$ does not have the stripe structure, because it has a random distribution of doping values, which are sorted following a Gaussian distribution.

tion theory applied to the LSCO series[13]. According to the CH results[19, 20], the square mesh phase separates into a bimodal distribution of charge. For underdoped samples the phase separation is essentially total and leads to two halves where the 7 stripes at the left are characterized by local doping $\rho(l) \approx 0$ and the 7 at the right side have $\rho(l) \approx 2\rho_m$ as showed in the top panel of Fig.(5) (for $\rho = 0.05$). As ρ_m increases, the charge fluctuations also increases, changing the properties of a compound into metallic, and superconducting at low temperatures. Thus, for compounds with $0.06 < \rho_m < 0.20$, at low temperatures, as one can see in Fig.(5), there are also stripes with non-vanishing densities at low density regions. Notice that the values of Δ are constant in a given stripe, in all its sites "i", and that is why we may plot $\Delta(i,T)$ at each site, although that is meaningful only at a given stripe $\Delta(l,T)$. As the density of charge carriers increases, it produces the $\Delta(i,T)$ at the very low doping sites, i.e., in the left region of Fig.(5). When this occurs at low temperatures, it is possible that the system becomes superconductor at $T_c(\rho_m)$ by the percolation of all the local superconducting regions[26, 27] and it can hold a dissipationless current. The percolation among the local regions where $\Delta(l,T)$ in non-zero may occur either directly or by Josephson coupling and the associated temperature, or

the superconducting temperature $T_c(\rho_m)$, is also shown in the panels of Fig.(5). Notice that the compound with $\rho = 0.22$ does not have the stripe structure, it has just a Gaussian distribution of doping values, since it is larger than the PST threshold of $\rho \approx 0.2$.

Above $T_c(\rho_m)$, depending on the value of ρ_m, the compounds may be formed by mixtures of superconducting, insulator and normal domains and above the pairing formation temperature (the onset temperature which sometimes is called the lower pseudogap), they are a disordered metal with mixtures of normal ($\rho(l) > 0.05$) and insulator ($\rho(l) \leq 0.05$) regions. For $\rho_m \geq 0.20$ the charge disorder is practically zero, with a small fluctuation around ρ_m. From these calculations, we identify $T_{onset}(\rho_m)$ as the highest temperature ($T_c(l)$) which induces a $\Delta(l,T)$ in a given compound which is easily seen in the panels of Fig.(5). T_{onset} may be also identified with the onset of Nernst signal[13]. To make clear how the values of T_{onset} of a given compound are obtained, we show the larger values of $T_c(i)$ and $T_c(l)$ in Fig.(6) for the sample with $\rho = 0.15$.
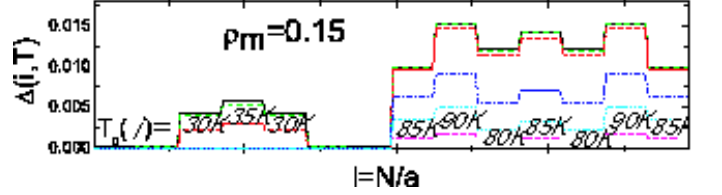

FIG. 6: (Color on line) To explain the concept of local superconducting temperature and specially how certain regions develops a non-vanishing pairing amplitude, we plot the $\Delta(i,T)$ or $\Delta(l,T)$ for each site "i" of our two dimensional 14×14 array. The onset temperatures values for which these pairing amplitudes develop for each stripe are clearly indicated in the figure, and the highest value, namely, $T = 90K$, is taken as the lower pseudogap temperature of this compound.

V. H_{c2} RESULTS AND CALCULATIONS

It is well known that the Ginzburg-Landau (GL) upper critical field of a homogeneous superconductor is a linear function of the temperature near T_c and falls to zero at this temperature[28]. This behavior is not observed by our measurements, showing another departure from conventional properties. As we can see in Fig.(7), the $H_{c2}(T)$ experimental points for both samples are linear only near and below $T/T_c \leq 0.9$. As the temperature increases it performs an upturn curvature falling to zero further beyond T_c. Consequently, we need some new ideas or theories to interpret these results.

From these $H_{c2}(T)$ curves, we can estimate that the upper critical field goes to zero at about 31.1 K, which is substantially higher than the value $T_c = 22.8K$ of the 8 %-Sr sample (from resistivity measurements as discussed in section II). Similarly, $H_{c2}(T)$ falls to zero at 24.1K for

the 25 %-Sr sample which has a $T_c = 19.9$K. Thus we see that $H_{c2}(T)$ vanishes at temperatures 8-22% larger than T_c. This effect, the nonzero value of H_{c2} above T_c, is quite unusual for a normal superconductor but it was also observed in LSCO and Bi-2201 cuprates by the group of Wang et al[29]. We will show below that this result may be explained as a consequence of the intrinsic disorder in HTSC, namely the presence of regions with different local dopings and distinct superconducting local temperatures $T_c(l)$.

In order to provide an interpretation to these results, we applied a generalization of the GL $H_{c2}(T)$ expression following along the lines described by Caixeiro et al[28]: The GL upper critical field near T_c of a homogeneous superconductor may be written as

$$H_{c2}(T) = \frac{\Phi_0}{2\pi\xi_{ab}^2(0)}\left(\frac{T_c - T}{T_c}\right). \quad (T < T_c) \quad (1)$$

At a temperature T, we take each superconducting region in the sample characterized by a $\rho(l)$ as the source that generates a $\Delta(l)$ to produce a magnetic response and displays a local H_{c2}, provided that $T \leq T_c(l)$. As discussed, these regions can be in stripe or others forms, but the important point is that they are characterized by a region of fairly constant density $\rho(l)$ that at $T \leq T_c(l)$ may shield the applied magnetic field. Thus each of such given local region has a local superconducting temperature $T_c(l)$ and will contribute to the upper critical field with a local linear upper critical field $H_{c2}^l(T)$ near $T_c(l)$ according to the usual GL approach. This is justified because each region has a constant density, like a type II low temperature superconductor, and should posses its own H_{c2} that is expected to vanish linearly at $T_c(l)$.

Consequently the total contribution of the local superconducting regions to the whole sample upper critical field is the sum of all the $H_{c2}^l(T)$'s,

$$H_{c2}(T) = \frac{\Phi_0}{2\pi\xi_{ab}^2(0)}\frac{1}{W}\sum_{l=1}^{W}\left(\frac{T_c(l) - T}{T_c(l)}\right)$$

$$= \frac{1}{W}\sum_{l=1}^{W} H_{c2}^l(T). \quad (T < T_c(l) \leq T_{onset}(\rho_m)) \quad (2)$$

Where W is the total number of superconducting regions, stripes or islands with its local $T_c(l) \geq T$. The maximum value of $T_c(l)$ is the pseudogap temperature identified above as the $T_{onset}(\rho_m)$. For the LSCO series a coherence length of $\xi_{ab}(0) \approx 22\mathring{A}$ is used, in accordance with the measurements[28]. This value of $\xi_{ab}(0)$ leads to $H_{c2}(0)=\Phi/2\pi\xi_{ab}^2(0)=64$T. Due to the limitations of the GL approach, we expect the result of this equation to be accurate only near and above the system T_c.

Fig.(7a) shows both the H_{c2} results of the generalized GL calculations together with the experimental values for underdoped $La_{1.92}Sr_{0.08}CuO_4$ compound. In the calculations on this compound, we used a maximum temperature of superconducting formation, $T_{onset} \approx 90K$
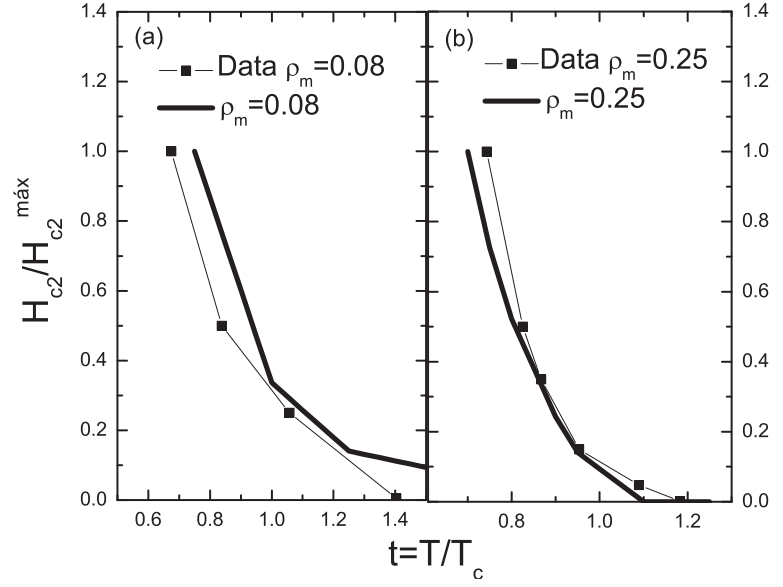

FIG. 7: Experimental points (connected by a thin line) against the reduced temperature $t = T/Tc$ and the calculated curve (thick line) of H_{c2} considering inhomogeneous samples with a stripe distribution of local superconducting temperature $T_c(l)$ for $\rho = 0.08$ and a similar calculation with a Gaussian distribution for $\rho = 0.25$.

(the maximum $T_c(l)$ for this sample) from pseudogap estimates[5] and from our calculations shown in Fig.(5). This is the reason why the calculated curve falls to zero at large values of the reduced temperature $t = T/T_c$. The measured critical temperature is, by the first derivative of the resistivity, $T_c(\rho_m) = 22.8$K. Thus, *using no adjustable parameters*, only values taken from experiments, we are able to obtain very reasonable agreement with the H_{c2} experimental values and, more importantly, a clear explanation why it does not vanish at T_c: at temperatures just above T_c there are some superconducting islands that do not percolate, leading to a finite resistivity, but they are still large enough to produce a clear magnetic response. The sum of such local magnetic responses is clearly seen in our experiments and in other H_{c2} measurements[29]. The magnetic contributions from non percolated islands above T_c was also measured in the form of an anomalous magnetization[23, 30] for underdoped compounds. Such results were interpreted within the framework of the critical state model on a charge disordered superconductor made up of islands[31], very close to the above approach.

Fig.(7b) also shows the data and the calculations on the overdoped $La_{1.75}Sr_{0.25}CuO_4$. Perhaps the PST line or upper pseudogap vanishes at $\rho_m = 0.20$[2], what is in agreement with many experiments that indicate more ordered behavior to overdoped than the underdoped compounds[13, 32]. Thus, we considered just a small Gaussian variation in the local charge density which yielded also a small variation in $\Delta T_c(\rho_m) \approx 18\%$. This calculation, with small fluctuations instead of large stripe

like variations, is in agreement with the Fermi liquid behavior of overdoped samples. Accordingly, the variations of $\Delta T_c(l)$ are very similar to the compound with $\rho_m = 0.22$, showed in the last panel of Fig.(5). As a consequence, the calculated $H_{c2}(T)$ curve of the overdoped sample falls to zero just 18% above T_c, while the underdoped vanishes at a much larger temperature.

VI. CONCLUSION

We observed that the measured $H_{c2}(T)$ curves for both underdoped and overdoped $La_{2-x}Sr_xCuO_4$ compounds display several non-conventional features like the positive curvature and non-vanishing values above T_c.

The original GL approach to H_{c2} near T_c fails to reproduce these behaviors. However it is possible to describe qualitatively well the observed behavior by a generalization of the GL theory that takes into account the intrinsic charge inhomogeneities. As an additional step towards an unified description, we assumed that such disorder was originated from a phase separation transition, possibly near the upper pseudogap temperature. The calculations were done in connection with the BdG formalism to obtain the distribution of local superconducting temperature $T_c(l)$, that is the onset of local pairing amplitude, on clusters with local charge density $\rho(l)$.

The measurements yield stronger non-conventional behavior for the underdoped sample which may be an indication of a larger degree of inhomogeneity, in agreement with other experiments[8, 32]. The different degrees of disorder were taken into account by the phase separation and this unified approach reproduced well the H_{c2} results. Thus we conclude that the observed unusual features associated with H_{c2} for both samples are consistent with the presence of charge inhomogeneities in the $La_{2-x}Sr_xCuO_4$, which depending on the value of x or ρ_m, appear either in the form of stripes or in the form of small fluctuation around the average doping level.

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